# CAUSES OF THE APPARENT HEAT TRANSFER **DEGRADATION FOR REFRIGERANT MIXTURES**

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### ABSTRACT

This paper presents an investigation into the causes of the apparent heat transfer degradation associated with horizontal-annular flow evaporation of refrigerant mixtures. The apparent heat transfer degradation is the difference between the measured heat transfer coefficient and the heat transfer coefficient that would be obtained from a linear interpolation of the single component values. The degradation is apparent since the linearly interpolated values have no physical basis. For horizontal-annular flow evaporation, most of the heat transfer degradation is a consequence of the use of the locally uniform equilibrium temperature in the measurement and calculation of the heat transfer coefficient. In reality, both circumferential and radial composition gradients can exist within the liquid film which cause temperature distributions that deviate significantly from a uniform saturation temperature. If the actual liquid-vapor interface temperatures (local vapor temperatures) were used in the calculation of the measured heat transfer coefficient for the impose heat flux condition, most of the apparent degradation would not exist. The remainder of the heat transfer degradation is due to nonlinear mixture property effects. Previously published measured heat transfer coefficients for three mixtures were investigated. The focus of the study was to determine the magnitude and the cause of the individual components of the heat transfer degradation of the studied mixtures.

### NOMENCLATURE

### English symbols

specific heat (kJ/kg-K) thermal conductivity (W/m-K)  $h_i$ h<sub>26</sub> from linear interpolation of single components (W/m<sup>2</sup>-K) h<sub>26</sub> predicted using single component correlation (W/m<sup>2</sup>-K) two-phase heat transfer coefficient (W/m<sup>2</sup>-K) h<sub>20</sub> m P P q Q r T<sub>i</sub> T<sub>n</sub> mass flow rate (kg/s) absolute pressure (Pa) critical pressure (Pa) reduced pressure, P/Pc heat flux (W/m<sup>2</sup>) mass flux (kg/m<sup>2</sup>-s) coordinate perpendicular to heat transfer surface (m) temperature of liquid-vapor interface (K)

saturated fluid temperature (K)

inside tube wall temperature (K) mole fraction of more volatile component mass fraction of more volatile component

x<sub>m</sub> thermodynamic quality

coordinate along heated surface (m)

### Greek symbols

T.

 $h_i - h_{2\phi} (W/m^2-K)$  $\Delta h_{2\phi}$ density (kg/m<sup>3</sup>)

experimentally measured density (kg/m<sup>3</sup>)  $\rho_{\rm exp}$ 

viscosity (kg/m-s)

# Subscripts

bottom of tube

liquid

mixture or mass

top of tube t

vapor

1 component number one

component number two

### INTRODUCTION

Pool boiling of mixtures has been practiced since antiquity. The ancient Greeks made their drinking water by distilling sea water. In the 19th century oil was refined by distillation to make kerosene for lamps. Aithough the practical application of mixture heat transfer is very old, the experimental and theoretical study of it is relatively new. The study of intube flow boiling of refrigerant mixtures is especially recent. During this short period of study, researchers have found that liquid mixtures do not evaporate as efficiently as single component liquids. However, mixtures of refrigerants can be used to enhance the efficiency of refrigeration equipment as compared to single component refrigerants (Mulroy et al., 1988). Unfortunately, the decrease or degradation in the efficiency of the evaporation of mixtures increases the costs of the performance improvements that can be achieved by using mixtures in cycles. This study is an attempt to further the understanding of horizontal flow boiling of mixtures with the hope of generating ideas that might lead to reduction of the heat transfer degradation associated with refrigerant mixtures.

There are two fundamental thermodynamic differences between single component fluids and mixtures which, in turn, cause fundamental differences between the phase change characteristics of single component fluids and mixtures. First, at constant pressure, the mixture temperature rises during evaporation, while the temperature of the single component fluids remains constant. Second, the liquid and vapor compositions are different in the mixture, while they are identical in the single component. These points are demonstrated by the phase equilibrium diagrams for the three binary mixtures investigated here, i.e., R22/R114, R12/R152a, and R13B1/R152a which are shown in Fig.s 1 through 3, respectively.

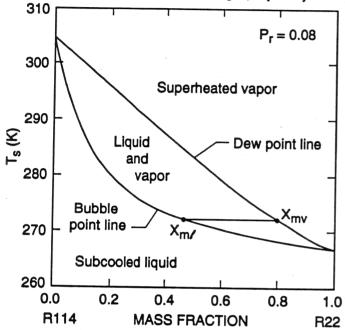


Fig. 1 Phase equilibrium diagram for R22/R114 at  $P_r = 0.08$ 

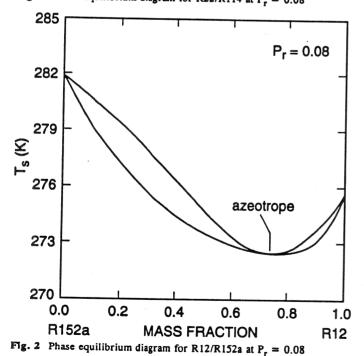


Figure 1, the phase equilibrium diagram for the R22/R114 mixture at a reduced pressure  $(P_r)$  of 0.08, represents the thermodynamic state of the mixture at equilibrium conditions. The phase equilibrium diagram is a

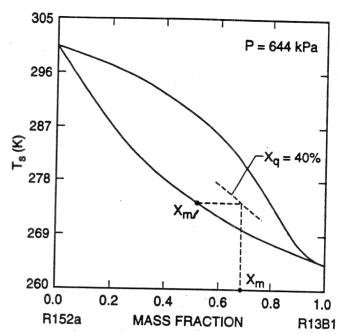


Fig. 3 Phase equilibrium diagram for R13B1/R152a at P = 644 kPa

plot of equilibrium temperatures versus the mass fraction of the more volatile component. The more volatile component is R22, since its equilibrium or saturation temperature is lower than that of R114 at the same pressure. The lower line, the bubble point line, represents the variation of the liquid saturation temperature with composition. The upper line, the dew point line, represents the variation of the saturated vapor temperature with composition. The area between the dew point and bubble point lines represents a two-phase mixture with a liquid of composition  $x_{ml}$  and a vapor of composition  $x_{mv}$  in coexistence. The distance between the dew point and bubble point lines, i.e., the temperature glide, may loosely be used to determine the potential for the heat transfer degradation of a particular mixture. The heat transfer of a binary mixture, which has a large temperature glide, is likely to be lessened by concentration gradients. Correspondingly, an azeotrope, which is a mixture that has no temperature glide, is likely to exhibit very little heat transfer degradation. Figure 2 shows that an azeotrope exists at  $x_m = 73.8\%$  R12 as depicted by the intersection of the dew and bubble

Figure 4 illustrates the concentration gradients that are established within a liquid mixture film as a consequence of preferential evaporation of the more volatile component. Notice that there are concentration gradients in two directions: (1) parallel to the uniformly heated surface  $(dx_{ml}/dy)$ , and (2) perpendicular to (or radially from) the uniformly heated surface  $(dx_{ml}/dr)$ .

The concentration gradient parallel to the heated surface shown in Fig. 4 is induced by the constant heat flux boundary condition and the varying film thickness. As a mixture evaporates, the bulk fluid is depleted of the more volatile component which reduces the mass fraction of the mixture. The mass fraction coordinate is measured along the wall, increasing in the vertical y-direction. The mass fraction of the thin-film  $(x_{mlb})$  region is less than that of the thick-film  $(x_{mlb})$  region since the thin-film region, having less mass, is further along the two-phase spindle of the equilibrium diagram. Consequently, a film thickness gradient has induced a concentration gradient along the heated surface. Since gravity imposes a nonuniform circumferential film thickness distribution for horizontal annular flow, the above argument describes the mechanism of the circumferential concentration gradients for annular flow within a horizontal tube. The temperature of the liquid-vapor interface at the top of the tube

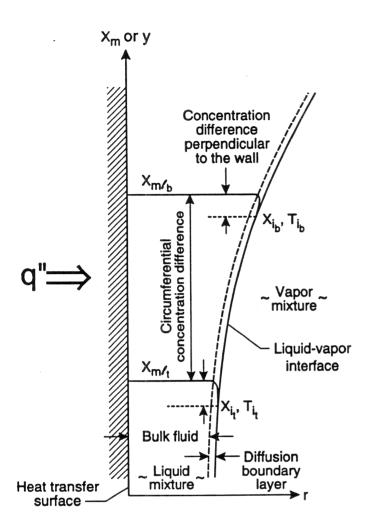


Fig. 4 Concentration gradients within the liquid film

 $(T_{it})$  is greater than that at the bottom of the tube  $(T_{ib})$ . The magnitude of the liquid-vapor interface temperatures are determined by both the circumferential and the radial concentration gradients.

The concentration gradient perpendicular to the heated surface exists primarily within the thickness of the diffusion boundary layer at the liquid-vapor interface, as shown in Fig. 4. Turbulent mixing prevents the formation of concentration gradients within the bulk liquid. In summary, evaporation depletes the diffusion boundary layer of the more volatile component and the convection confines the concentration gradient to a narrow region within or close to the liquid-vapor interface. This describes the mechanism of the radial concentration gradients for annular flow within a horizontal tube.

Figure 5 demonstrates the variation of the two-phase heat transfer coefficient with respect to composition for an illustrative mixture. The dashed line represents experimental data. Notice that the heat transfer coefficient can be less than that for either pure component. The solid line,  $h_i$ , is a linear interpolation between the heat transfer coefficients of the pure components. The straight line has no physical meaning; however, it is used as a reference from which the degradation of the heat transfer coefficient of binary mixtures can be quantified. The heat transfer degradation ( $\Delta h_{2\phi}$ ) is the difference between the interpolated heat transfer coefficient ( $h_i$ ) and the measured heat transfer coefficient ( $h_{2\phi}$ ) for a given composition.

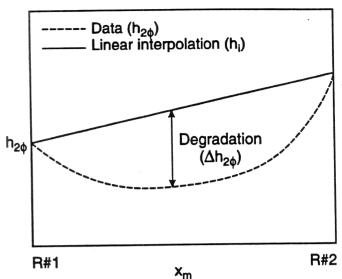


Fig. 5 Typical relationship of mixture flow boiling with respect to composition

Several explanations for the heat transfer degradation associated with mixtures have been postulated. Two popular reasons are the loss of available superheat and mass transfer resistance. Also, Ross et al. (1987), Jung and Didion (1989) and Stephan and Preusser (1979) attribute a portion of the degradation to the nonlinear variation of the thermodynamic and transport properties of mixtures with composition. The nonlinear variation of properties with respect to composition may contribute to a nonlinear degradation of the heat transfer coefficient. Stephan and Korner (1969) suggest still another reason for the degradation: mixtures must produce more work to form bubbles than for an equivalent pure fluid.

Loss of available superheat, as described by Shock (1982), is the loss of the heat transfer driving potential due to the increase of the fluid temperature upon evaporation. For a fixed tube-wall temperature, an increase in the fluid temperature results in a reduction or a loss of temperature difference between the wall and the fluid. The data examined here are for a constant, imposed heat flux boundary condition, not a constant wall temperature boundary condition. For the constant heat flux case, the heat transfer driving potential is the heat flux, which dictates the tube-wall temperature. A loss in driving potential cannot be imposed by an increase in fluid temperature upon evaporation since the wall temperature will rise, as the fluid temperature rises, to satisfy the imposed heat flux boundary condition. For the above reasons, the argument for the loss of available superheat does not strictly apply to the constant heat flux boundary condition and, consequently, cannot be investigated in this paper.

Mass transfer, as defined in this paper, is the movement of a liquid component due solely to a concentration gradient. The motion of the liquid is induced by the tendency of the liquid to achieve a uniform equilibrium concentration. The magnitude of the mass flux, due to mass transfer, is insignificant compared to that due to evaporation by heat exchange. Therefore, mass transfer cannot significantly reduce the heat transfer by a movement of fluid which is opposed to the evaporation. However, mass transfer can indirectly effect the heat transfer by determining the magnitude of the concentration gradients. In turn, the concentration gradients establish the temperature distributions which control the heat transfer. In summary, the mass transfer effects the heat transfer coefficient primarily by altering the temperature through concentration gradients and not by the movement of fluid.

Mass transfer resistance is defined in this paper as a resistance to the neutralization of concentration gradients. The mass transfer resistance indirectly causes a degradation in the heat transfer by raising the liquid

temperatures. For clarity, concentration gradients rather than mass transfer resistance is used to discuss the heat transfer degradation.

The additional work of bubble formation cannot be studied in the convective region. All of the data which were examined are in the convective, evaporative flow region. Nucleate boiling is suppressed in the convective region (Chen, 1966). Therefore, the additional resistance due to bubble formation in a mixture cannot be examined using the cited data.

For the above reasons, the focus of the investigation is on the effects of concentration gradients and nonlinear mixture properties on the heat transfer degradation. It cannot be known with any certainty that the above two effects are the only effects that contribute to the heat transfer degradation. However, it is assumed that the heat transfer degradation that is not due to the fluid property effect is due to concentration gradients.

For evaporative flow, it is speculated that most of the heat transfer degradation associated with concentration gradients results from the use of the saturated equilibrium temperature in the calculation of the heat transfer coefficient. If concentration gradients are present in the liquid, the actual liquid-vapor interface temperature (vapor temperature) will be greater than the saturated equilibrium temperature which is obtained from an overall energy balance and the measured pressure. If the actual liquid-vapor interface temperature was used to calculate the measured heat transfer coefficient the heat transfer coefficient would be greater than that calculated from the equilibrium temperature. Consequently, a large portion (possibly all) of the heat transfer degradation associated with concentration gradients can be attributed to the use of the equilibrium temperature in the calculation of the measured heat transfer coefficient.

### **EXPERIMENTAL WORK INVESTIGATED**

Only binary-mixture, horizontal-flow boiling with a constant tube wall heat flux is considered here (Ross, 1985 or Ross et al., 1987, and Jung and Didion, 1989 or Jung et al., 1989). The local two-phase heat transfer coefficient ( $h_{2\phi}$ ) was calculated as:

$$h_{2\phi} = \frac{q''}{T_{\psi} - T_{\phi}} \tag{1}$$

where  $q^*$  is the heat flux at the outside wall,  $T_w$  is the inside wall temperature, and  $T_s$  is the saturated fluid temperature evaluated at the measured pressure. Local wall temperature measurements were made along the tube length. The tube wall temperature was measured at four circumferential positions, 90 degrees apart, at each axial position along the tube. Four circumferential heat transfer coefficients (top, bottom, right, and left side) were calculated and averaged to obtain the value for the heat transfer coefficient at the given axial position.

Ross et al. (1987) have measured the local flow boiling heat transfer coefficient for various compositions of the R13B1/R152a mixture. Figure 6 presents the measured two-phase heat transfer coefficient  $(h_{2\phi})$  versus the mass fraction of the more volatile component R13B1. The data were taken for a saturation temperature, at the exit of the test section, of 270 K, a mass flux of 460 kg/(m<sup>2</sup> s), a thermodynamic quality of 40%, and an incident heat flux of 30 kW/m<sup>2</sup>. The solid line is a linear interpolation between the heat transfer coefficients for the single components. The degradation as compared to the single component reference line is greatest (65% of  $h_i$ ) at  $x_m = 0.82$ .

Jung et al. (1989) measured the local flow boiling heat transfer coefficient for various compositions of the R12/R152a mixture and the R22/R114 mixture. His data for a reduced pressure ( $P_r$ ) of 0.08 at the exit of the test section, a constant imposed heat flux of 17 kW/m², a thermodynamic quality of 65%, and a mass flow rate (m) of 0.023 kg/s are shown in Fig.s 7 and 8. The maximum heat transfer degradation for the R22/R114 mixture is located at an overall mass composition of 0.61. Its magnitude is 2040 W/m²-K or 37% lower than the linear interpolation between the  $h_{2\phi}$  for the pure components.

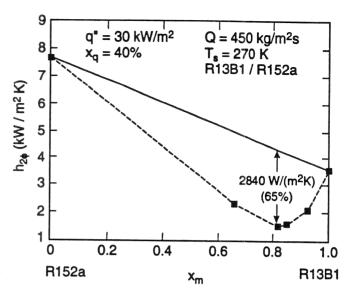


Fig. 6 Measured horizontal flow boiling heat transfer coefficients for the R13B1/R152a mixture (Ross et al. (1987))

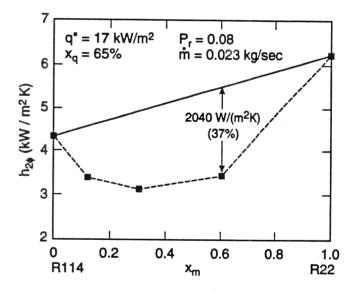


Fig. 7 Measured horizontal flow boiling heat transfer coefficients for the R22/R114 mixture (Jung and Didion, 1989)

The heat transfer degradation for the R22/R114 mixture is large, but not as large as that present for the R13B1/R152a mixture. It is intuitively reasonable to suggest that the large heat transfer degradation associated with the R13B1/R152a data is a result of the coupled effects of: (1) the large difference in mass concentration between the liquid and vapor phases  $(x_{mv} - x_{ml} = 0.12$  at  $x_m = 0.82$ ) and, (2) the relatively large molecular mass (149 g/mole) of the more volatile component (R13B1). The difference in concentration between the liquid and vapor phases represents the potential for concentration gradients within the liquid. The difference between the vapor and liquid composition  $(x_{mv} - x_{ml})$  for the R22/R114 mixture is approximately 0.02 mass fraction greater than that for the R13B1/R152a mixture from about 0.2 to 0.8 liquid mass fraction. Therefore, the potential for mass transfer resistance is slightly greater for the R22/R114 than it is for the R13B1/R152a mixture. However, the molecular mass of R22 is 86 g/mole, and that of R13B1 is 149 g/mole.

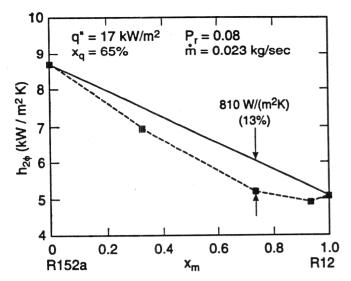


Fig. 8 Measured horizontal flow boiling heat transfer coefficients for the R12/R152a mixture (Jung and Didion, 1989)

In other words, the more volatile component of the R22/R114 mixture is lighter than that of the R13B1/R152a mixture. The speed at which molecules diffuse determines the magnitude of the concentration gradient (McCabe and Smith, 1976). For binary liquids, the rate of diffusion is primarily a function of: (1) the liquid viscosity, (2) the derivative of the log of the activity with respect to the log of the mole fraction of the more volatile component, and (3) the molecular mass of the components (Bird et al., 1960). The viscosity and the activity-composition data for the two mixtures do not differ significantly. If an analogy with vapor diffusion is permitted, then it is reasonable to assume that heavy liquid molecules, like those of R13B1, would diffuse more slowly than the lighter R22 liquid molecules. Consequently, there would be larger concentration gradients present in the R13B1/R152a mixture than in the R22/R114 mixture. Therefore, it is hypothesized that the heat transfer degradation of the R13B1/R152a mixture is larger than that of the R22/R114 mixture because the molecular mass of the more volatile component of the R13B1/152a mixture is greater than that of the R22/R114 mixture.

The phase equilibrium diagram for the R12/R152a mixture, in Fig. 2, shows that the maximum difference between the liquid and vapor compositions for that mixture is approximately 0.1 mole fraction. The fact that the composition difference is small suggests that the potential for heat transfer degradation should be small. The measured two-phase heat transfer coefficient for the R12/R152a mixture, shown in Fig. 8, satisfies the speculation by exhibiting only a 13% degradation in the heat transfer from the linear.

### EFFECT OF FLUID PROPERTIES ON ha

### Mixing Rules

The calculation of the heat transfer coefficient, as given by equation (1), requires relatively few fluid properties. However, its correlation and prediction rely heavily on the estimated or measured fluid properties. For this reason, it is essential that correlations are presented along with the fluid property mixing rules that were used to fit the data. The following analysis demonstrates the effect of the mixing rule on the determination of the two-phase heat transfer coefficient for mixtures in the convection-dominated regime.

The functional form of the Dittus-Boelter (1930) equation  $(k_1^{0.6}(c_{p_1}/\mu_1)^{0.4})\rho_1^{0.8}$  is frequently used to correlate the convection-dominated region of two-phase flow within a tube. The  $k_1$  is the thermal conductivity of the liquid;  $c_{p_1}$  is the specific heat of the liquid;  $\mu_1$  is the viscosity of the

liquid; and,  $\rho_1$  is the density of the liquid. These are the primary fluid properties which are necessary for the correlation and prediction of heat transfer coefficients for two-phase flow boiling.

An estimate of the fluid properties of a mixture can be obtained from the fluid properties of the pure components using mixing rules. Three typical mixing rules are: (1) linear, (2) ideal, and (3) non-ideal. The simplicity of the linear mixing or mass fraction averaging rules is attractive:

$$k_{1_{-}} = x_{m_1} k_{1_1} + (1 - x_{m_2}) k_{1_2}$$
 (2)

$$C_{p_{l_n}} = X_{\underline{n}_1} C_{p_{l_1}} + (1 - X_{\underline{n}_1}) C_{p_{l_2}}$$
 (3)

$$\mu_{I_m} = x_{n_1} \mu_{I_1} + (1 - x_{n_1}) \mu_{I_2}$$
 (4)

Linear mass fraction weighing mixing rules are seldom used to approximate the liquid thermal conductivity and the liquid viscosity. However, the linear mixing rule can be used to closely approximate the specific heat of a mixture.

The ideal mixing rules are slightly more complex than the linear mixing rules, but closely approximate the properties of a mixture where the pure components have similar vapor pressures and come from similar chemical families. The ideal mixing rules assume that there are no mixing effects that enhance or reduce the value of the property due to mixing (Reid et al., 1977).

$$k_{1_n} = \exp[x_{m_1} \ln(k_{1_1}) + (1 - x_{m_1}) \ln(k_{1_2})]$$
 (5)

$$\mu_{l_n} = \exp\left[x_{n_1} \ln(\mu_{l_1}) + (1 - x_{n_1}) \ln(\mu_{l_2})\right] \tag{6}$$

The non-ideal mixing rules chosen for this study have an additional term to account for the effects of mixing:

$$k_{l_n} = x_{n_1} k_{l_1} + (1 - x_{n_1}) k_{l_2} - 0.72 x_{n_1} (1 - x_{n_1}) |k_{l_1} - k_{l_2}|$$
 (7

$$\mu_{I_{s}} = \exp\left[x \ln(\mu_{I_{1}}) + (1 - x) \ln(\mu_{I_{2}})\right] + 0.85 \left[\rho_{I_{axp}} \left(\frac{x}{\rho_{I_{1}}} + \frac{(1 - x)}{\rho_{I_{2}}}\right) - 1\right] - 0.085$$
(8)

Equation (7) was obtained from Reid et al. (1977) and equation (8) was obtained from Jung and Didion (1990).

Figure 9 is used to examine the impact of the mixing rule on the prediction of the heat transfer coefficient for mixtures. The figure consists of four graphs of the predicted h<sub>24</sub> versus the mass fraction (x<sub>m</sub>) for the R22/R114 mixture. The uppermost line is the linear interpolation between the heat transfer coefficients of the single components, R22 and R114. The remaining three graphs are of the predicted flow boiling heat transfer coefficient (h<sub>p</sub>), using three different mixing rules to estimate the fluid properties used in the correlation. Jung's (1989) flow boiling correlation for single component fluids was used so that only the effect of the mixing rule on the heat transfer coefficient were examined. The predictions can be viewed as a heat transfer coefficient for the mixture if there were no concentration gradients present within the liquid.

Three general characteristics of Fig. 9 are evident. First, note that the predicted heat transfer coefficient is nonlinear with respect to the composition for all of the mixing rules. Second, the apparent heat transfer degradation is the greatest at a mass fraction of 70%, which is different from the mass fraction for the greatest value of  $x_{mv} - x_{ml}$ . This indicates that the nonlinear property effects are acting to minimize the heat transfer by a mechanism which is different from that of the concentration gradient effects. The consequence of the property effects interacting with the

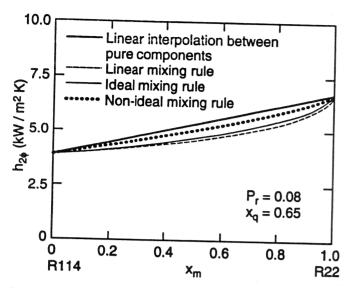


Fig. 9 Impact of the mixing rule on the apparent heat transfer degradation of the R22/R114 mixture

concentration gradient effects is to minimize the heat transfer at a composition which is a compromise between the two effects. Third, the apparent maximum heat transfer degradation becomes less as the mixing rule for the fluid property estimations progress from the linear to the ideal and finally to the non-ideal. In summary, the maximum deviation from the linear interpolation is 18% for the linear mixing rule, 14% for the ideal mixing rule, and 6% for the non-ideal mixing rule. Figure 9 demonstrates the importance of consistency in the use of mixing rules for estimating the fluid properties to be used in the heat transfer coefficient correlations.

# The Less Volatile Component

The removal of heat from the wall by convection for two-phase flow within a tube is governed by both the molecular conduction of heat through the liquid film and the transport of the liquid along the tube wall. Hence, the local properties of the liquid film determine the local rate of heat transfer. Figure 3 illustrates that the composition of the liquid increases in the less volatile component as the fluid evaporates. For this reason, care should be taken to evaluate the liquid properties at the liquid composition  $(x_m)$  (not the overall composition  $(x_m)$ ) in both the correlation and prediction of the flow boiling heat transfer coefficient with respect to flow quality  $(x_q)$ .

Figure 10 is a plot of the predicted heat transfer coefficient, using Jung's (1989) single component model, versus the overall composition. The figure shows that as large as a 15% error in the prediction of the heat transfer coefficient can occur if the fluid properties are evaluated at the overall composition rather than the liquid composition. The heat transfer coefficient for the R12/R152a mixture will be underestimated by using the overall composition to evaluate the liquid properties. Contrary to this, the heat transfer coefficient for the R22/R114 mixture will be overestimated by using the overall composition to evaluate the liquid properties.

In general, the liquid properties of a two-phase mixture more closely resemble those of the less volatile component than that which would be anticipated considering the overall composition. For a R22/R114 mixture, this results in heat transfer coefficients which are always below the linear interpolation of the heat transfer coefficients of the single components. However, the predicted heat transfer coefficients evaluated at  $x_{al}$  for the R12/152a mixture are slightly above the  $h_i$  values. The liquid properties of R152a are more beneficial for convection than the liquid properties of R12, resulting in an enhancement with respect to the overall composition. The opposite is true for the R22/R114 mixture where the liquid properties

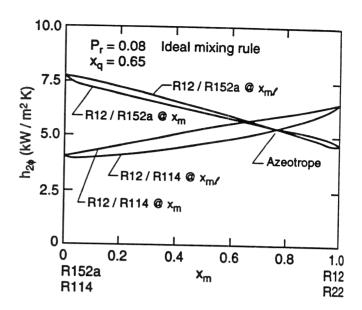


Fig. 10 Effect of evaluating liquid properties at  $x_m$  and  $x_{ml}$  on  $h_{2a}$ 

of the less volatile component do not promote the convection as well as the more volatile component. It may be possible to tailor a mixture which has heat transfer coefficients above the linearly interpolated values by selecting the less volatile component to have the best convective characteristics of all the components.

# COMPONENTS OF DEGRADATION

Following is an attempt to isolate and quantify the individual components of the total heat transfer degradation depicted in Fig. 5. The first section concentrates on determining the influences of fluid properties and liquid concentration gradients on the heat transfer coefficient. The last section attempts to isolate the proportions of the  $\Delta h_{24}$  that are due to the circumferential and radial concentration gradients.

In order to analyze the influence of fluid properties on the heat transfer coefficient in the absence of concentration gradients, the correlation for two-phase single component horizontal flow boiling by Jung and Didion (1989) was utilized. Figure 11 shows the two-phase heat transfer coefficient for the R12/R152a mixture, as predicted using the Jung and Didion (1989) correlation, versus the mass fraction of the more volatile component. The predicted values were adjusted to facilitate a fair analysis of the nonlinear property effects on the heat transfer. The difference between the prediction and the linear interpolation between the predicted single component heat transfer coefficients was transferred to Fig. 11 as a heat transfer degradation for the measured heat transfer coefficients due to nonlinear property effects. Figure 11 compares the experimental data to the adjusted predicted values for the R12/R152a mixture. The fluid properties degrade the heat transfer coefficient by 6% at  $x_m = 0.5$ . Concentration gradients are assumed to be responsible for the remaining 7% degradation in the heat transfer coefficient at  $x_m = 0.7$ .

Figure 12 demonstrates that both fluid properties and concentration gradients reduce the heat transfer coefficient for the R22/R114 mixture. Only a small portion, 14% of the apparent degradation  $(\Delta h_{2a})$  is due to fluid property effects of mixtures. The majority, 86% of the apparent degradation is due to concentration gradients within the liquid film. Both circumferential and radial concentration gradients contribute to the degradation due to concentration gradients. A closer look into the degradation due to the concentration gradients follows.

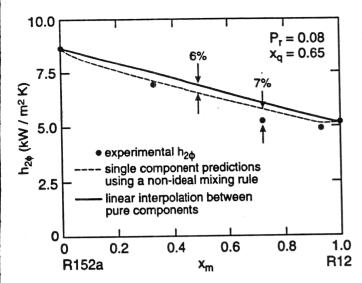


Fig. 11 Comparison of fluid property, and concentration gradient effects on the degradation of R12/R152a heat transfer

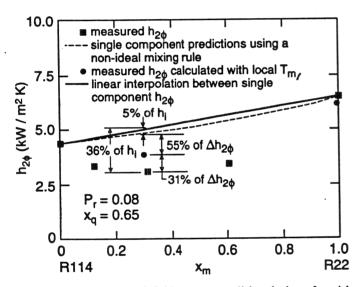


Fig. 12 Comparison of fluid property, radial and circumferential gradient effects on the degradation of R22/R114 heat transfer

Figure 13 is a plot of the heat transfer degradation ( $\Delta h_{2\phi}$ ) versus the difference between the vapor and liquid mass fractions (x<sub>mv</sub> - x<sub>ml</sub>). Recall that this difference represents the potential for concentration gradients within the liquid. Figure 13 shows that the heat transfer degradation of the R22/R114 data correlates well with the vapor-liquid composition difference, having a standard deviation of 1.5% from the straight line. Consequently, the heat transfer degradation appears to increase linearly with the difference in the liquid and vapor compositions. The linear rate of increase of the degradation with composition difference, for the R13B1/R152a mixture, is not as well defined by a straight line as it is for the R22/R114 mixture. The evidence for this is that the standard deviation of the  $\Delta h_{2\phi}$  from the linear least squares fit for the R13B1/R152a mixture is  $\pm 12\%$  which is much greater than that for the R22/R114 mixture. Notice that the heat transfer degradation for the R22/R114 mixture and the R13B1/R152a mixture have approximately the same rate of increase with respect to the increase in the difference in the vapor and liquid compositions (3.4 kW/m<sup>2</sup>-K-mass fraction difference). A given increase in the difference between the compositions of the phases results in the same net change in the heat transfer coefficient for both the R22/R114 and

the R13B1/R152a mixtures. Also, notice that the intercept of the R22/R114 mixture gives a degradation of  $0.3~\mathrm{kW/m^2}$ -K which is the degradation that would be expected in the absence of concentration gradients. Figure 9 confirms this hypothesis since the property effects alone cause nearly the same degradation over  $x_m = 0.4$  to 0.8. This suggests two postulations. First, the heat transfer degradation may be approximated as a sum of the degradation due to approximately constant property effects and that due to concentration gradients which are linearly dependent upon the difference between the vapor and liquid mass fractions. Second, the heat transfer degradation, due to the circumferential and radial concentration gradients, is directly related to the difference between the composition of the liquid and vapor phases.

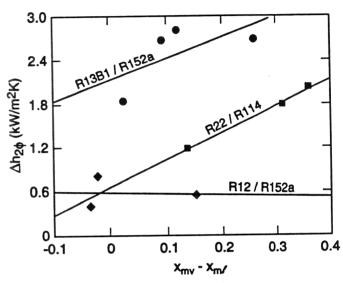


Fig. 13 Effect of vapor-liquid composition difference on the heat transfer degradation

The heat transfer degradation of the R12/R152a mixture appears to have little dependence upon the vapor-liquid mass fraction difference. This suggests that there are no liquid concentration gradients associated with the R12/R152a mixture since the degradation is not a function of the composition difference. Yet, the degradation is still greater than that due to the mixture property effects. The remaining apparent degradation may be a consequence of: (1) the uncertainty in the heat transfer coefficient measurements, and/or (2) the inability of the Jung and Didion (1989) model for the single component heat transfer coefficients to precisely predict the behavior of mixtures without concentration gradients.

Figure 14 is a graph of the difference in vapor and liquid mass fraction versus the liquid mass fraction for the three mixtures. The data symbols correspond to the liquid mass fraction at which the heat transfer coefficients of Fig.s 6 through 8 were measured. The vapor-liquid mass fraction difference for the R22/R114 mixture is the largest of the three mixtures peaking at near 0.4 mass fraction difference. The vapor-liquid mass fraction difference for the R13B1/R152a mixture is nearly the silhouette of the R22/R114 mixture, but is approximately 0.02 lower than that for R22/R114 over most of the liquid mass fraction range.

If it is assumed that the largest  $x_{mv}$  -  $x_{ml}$  will produce the greatest heat transfer degradation, then Fig. 14 shows that this occurs at a liquid composition of approximately 0.28 for both the R22/R114 and the R13B1/R152a mixture. The vapor-liquid composition difference associated with the maximum degradation lies between the vapor-liquid composition difference for the measured points for the R22/R114 mixture. Therefore, the maximum R22/R114 degradation was probably revealed by the measured heat transfer coefficients, shown in Fig. 7. However, the liquid compositions, for which data were taken for the R13B1/R152a mixture,

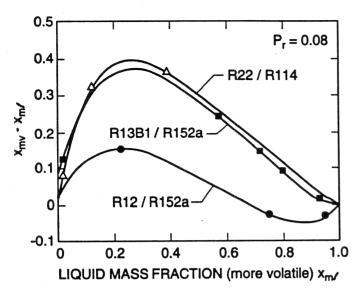


Fig. 14 Concentration difference versus liquid composition

are all above the composition that would demonstrate the largest possible heat transfer degradation. Figure 14 shows that at  $x_{\rm ml}=0.53$ , the potential for concentration gradients is 35% below its maximum value. Consequently, if the heat transfer coefficient was measured at a liquid mass fraction of 0.28, the heat transfer degradation may have been substantially greater than that which was measured at a mass fraction of 0.82.

Oddly, the R12/R152a data is inconsistent with the above trends. The maximum, measured heat transfer degradation of the R12/R152a mixture corresponds to the azeotropic composition of the mixture. The degradation associated with the R12/R152a mixture is of the same magnitude as the accuracy of the heat transfer coefficient measurement (± 10%). Consequently, the composition at which the actual maximum heat transfer degradation occurs cannot be known with much confidence.

### CIRCUMFERENTIAL AND RADIAL GRADIENTS

Ross et al. (1987) had hypothesized the existence of a circumferential liquid concentration gradient for evaporating mixtures. Jung et al. (1989) validated this speculation by taking concentration samples from the top, the bottom, and the side of the tube. Figure 15 shows that the measured concentration variation for the R22/R114 mixture, at an overall composition of 48% mole R22, is as large as 0.07 mole fraction from the top to the bottom of the tube. Jung's measurement was probably not of the liquid-vapor interface composition. Most likely, the composition that was measured was that of the bulk liquid, as shown in Fig. 4.

When the heat transfer coefficient is calculated, the intention is to define it across the actual temperature difference for which the heat transfer is occurring: the temperature of the heated surface minus the temperature of the liquid-vapor interface ( $T_w$  -  $T_i$ ). The temperature of the liquid-vapor interface (i.e, the vapor temperature), in the Ross et al. (1987) and the Jung et al. (1989) studies, was assumed to be circumferentially uniform and equal to the equilibrium temperature. The equilibrium or saturation temperature was determined from the measured vapor pressure, the measured overall composition, the calculated thermodynamic quality and The saturation temperature will be an equation of state (EOS). circumferentially uniform only if the composition of the bulk liquid is uniform around the circumference of the tube. Jung et al. (1989) have shown that a circumferentially uniform liquid composition cannot be guaranteed, which implies that the liquid-vapor interface temperature can also vary. The importance of the circumferential variation is evident in the calculation of the heat transfer coefficient for a particular axial

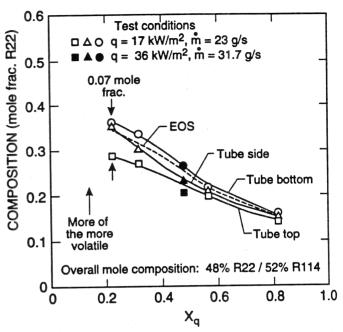


Fig. 15 Measured circumferential liquid film concentration distribution for a 48% mole R22/R114 (Jung et al., 1989)

position, which is obtained by averaging the heat transfer coefficients for the four positions around the circumference of the tube. Thus, the calculation of the heat transfer coefficient using the circumferentially uniform saturation temperature assumption may be inaccurate for some annular flow patterns.

Ideally, both the radial and circumferential concentration gradients should be considered in the evaluation of the liquid-vapor interface temperature for the calculation of the local heat transfer coefficient. Since Jung et al. (1989) circumferentially measured the composition of the bulk liquid, they were able to calculate the circumferential variation of the local saturation temperature for that composition. This temperature still does not represent the temperature of the liquid-vapor interface since: (1) there is a further drop in composition from the bulk of the liquid to the liquid-vapor interface, and (2) the bulk of the liquid will be superheated. The liquid-vapor interface is the only portion of the liquid which is possibly in thermodynamic equilibrium. The liquid-vapor interface temperature is, most likely, equivalent to the equilibrium temperature at the composition of the interface. Therefore, both radial and circumferential concentration gradients influence the temperature distribution of the liquid-vapor interface.

Jung et al. (1989) calculated the local heat transfer coefficient based on the equilibrium temperature obtained from the circumferentially varying composition of the bulk liquid (xml). The circumferentially averaged local heat transfer coefficient, if calculated using the circumferentially varying saturation temperature, is higher than that calculated using the uniform saturation temperature obtained from the thermodynamic quality and pressure. Figure 12 shows the value of the  $h_{24}$  (at  $x_m = 0.31$ ) if it is calculated using the circumferentially varying saturation temperature. The "corrected" heat transfer coefficient is 18% greater than that calculated using the uniform saturation temperature. Consequently, the apparent heat transfer degradation is not as large as the uniform saturation temperature calculation method suggests. In fact, 35% of the apparent degradation due to concentration gradients is due to the use of a circumferentially uniform saturation temperature in the calculation of the heat transfer coefficient. It is speculated that the remaining degradation due to concentration gradients is caused by the radial concentration gradients within the liquid film. In summary, for the heat transfer conditions shown in Fig. 12 at (x<sub>n</sub>

= 0.31), 14% of the heat transfer degradation is due to the nonlinear property effects, 31% is due to the circumferential concentration gradients, and 55% is due to the radial concentration gradients. Therefore, it is likely that, if the actual liquid-vapor interface temperature was used to calculate the heat transfer coefficient, the degradation due to nonlinear property effects would be the only degradation present.

Since a limited amount of data are presented, it cannot be assumed that the circumferential averaged heat transfer coefficient calculated using the composition of the bulk liquid will always be higher than that using the uniform equilibrium temperature. However, an argument can be made to establish that, in general, the above trend should be true for most horizontal-annular flow evaporation with a constant heat flux boundary condition. The phenomenon is a result of the nonuniform liquid film thickness around the circumference of the tube, i.e., a thick-film region in the bottom half of the tube and a thin-film region near the top half of the tube. The velocities of the tube-top and tube-bottom liquid films are nearly the same, due to the imposed axial pressure gradient of the vapor phase. Thus, the liquid mass flow rate of the tube-top is less (possibly much less) than that for the bottom of the tube. The liquid stream with the smaller mass flow rate will progress from liquid to vapor sooner than the stream with the larger mass flow rate for the identical heat input (uniform heat flux boundary condition). The temperature and quality of the mixture increases as it evaporates. For this reason, the liquid in the thin-film region will always be at a higher temperature than the liquid in the thickfilm region. Also, the liquid near the top of the tube (the thin-film region) will be at a higher temperature than that for a flow with a circumferentially uniform film thickness distribution. For this reason, the heat transfer coefficient at the top of the tube, using the local liquid temperature, will always be higher than that using the uniform equilibrium temperature. Likewise, the heat transfer coefficient at the bottom of the tube calculated from the local liquid temperature will always be marginally lower than that calculated using the uniform equilibrium temperature. But, the top of the tube will have a significantly higher heat transfer coefficient compared to the uniform method and the bottom of the tube will have a marginally lower heat transfer coefficient compared to the same method. Consequently, the average of the top and bottom heat transfer coefficients, using the actual liquid-vapor interface temperature, will tend toward being larger than that calculated from the uniform equilibrium temperature.

### CONCLUSIONS

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Several precautions must be used when predicting and correlating twophase heat transfer data for mixtures. First, non-ideal mixing rules which most closely approximate the behavior of the mixtures should be used to correlate the data. The presentation of a new correlation should include the mixing rules that were used to generate it. The liquid composition and not the overall composition of the mixture should be used to evaluate the local fluid properties since it is the liquid film which locally controls the heat transfer.

The two-phase heat transfer coefficient predicted using the linear, the ideal, and the non-ideal mixing rules all resulted in a nonlinear variation of the heat transfer coefficient with the mass fraction. The heat transfer coefficients predicted using the non-ideal mixing rule were closest to the linear interpolation of the heat transfer coefficient between the single component fluids.

The degradation of the heat transfer coefficient by concentration gradient effects appears to be more significant for mixtures of fluids having widely different boiling points. The R22/R114 mixture has a maximum mass concentration difference which is more than double that of the R12/R152a mixture. The heat transfer degradation for the R12/R152a mixture is consequently much smaller than that for the R22/R114 mixture. For equal differences between the vapor and liquid mass fractions, concentration gradient effects appear to be more significant where the more volatile component has a relatively high molecular mass. For example, the R22/R114 and the R13B1/R152a mixtures have approximately the same

concentration difference between the liquid and the vapor. But the R13B1 molecule is heavier than the R22 molecule and consequently the R13B1/R152a mixture has a larger heat transfer degradation. In summary, it is speculated that the  $x_{mv} - x_{ml}$  is the potential for concentration gradients and the molecular mass of the more volatile component determines the magnitude of the concentration gradient which in turn determines the magnitude of the heat transfer degradation.

The largest measured heat transfer degradation, 2840 W/m²-K, of all the studied mixtures was for the R13B1/R152a mixture. The difference between the vapor and the liquid mass fraction associated with this data point was 0.13 which was 35% below its maximum value. Consequently, a heat transfer degradation greater than that which was measured may be expected at a mass fraction of 0.53 since this composition corresponds to a vapor-liquid composition difference of 0.28.

An argument can be made to establish that the heat transfer coefficient calculated accounting for the circumferentially varying saturation temperature will be higher than that calculated using a circumferentially uniform saturation temperature for most horizontal-annular flow evaporation. The R22/R114 heat transfer coefficient, for one particular set of conditions, calculated using the varying saturation temperature was 18% greater than that calculated using a uniform saturation temperature.

Typically, the measured flow boiling heat transfer coefficient is calculated using a uniform equilibrium temperature. The apparent heat transfer degradation for this situation can be attributed to three phenomena: (1) fluid property effects, (2) radial liquid concentration gradients, and (3) circumferential concentration gradients. For the R22/R114 mixture, 14% of the heat transfer degradation is due to fluid property effects, 55% is due to radial concentration gradients, and the remaining 31% is attributed to the circumferential concentration gradient. It is postulated that, if the actual liquid-vapor interface temperature were used to calculate the measured heat transfer coefficient it would be greater than that calculated from the equilibrium temperature.

Although concentration gradients within the liquid film tend to degrade the heat transfer, nonlinear fluid property effects may act to enhance the heat transfer. This can occur when the heat transfer characteristics of the less volatile component are more favorable for high heat transfer rates than those of the more volatile component. The heat transfer performance more closely resembles that of the less volatile component. Thus, a mixture could be tailored to minimize the heat transfer degradation by selecting a mixture such that the heat transfer coefficient of the less volatile component is greater than that of the more volatile component.

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